

PATENT SPECIFICATION

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(54) CARBON ELECTRODES

(71) We, NIPPON CARBON CO., LTD., of 6-1, 2-chome, Hatchobori, Chuo-ku, Tokyo, Japan, a Japanese body corporate, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

10 This invention relates to carbon electrodes and more particularly to high-strength and spalling-resistant carbon electrodes for electrical furnaces wherein iron, steel and alloys are manufactured.

15 The term "carbon electrode" used herein is intended to mean non-graphitized and graphitized carbon electrodes including the electrode bodies and nipples.

20 It is well known that artificial graphite electrodes are used in electric furnaces. However, the technique of operating electric furnaces and the technique of producing artificial graphite have each been improved, and these improved techniques have had effects on each other. With the recent increase of electric furnaces in scale and capacity, higher electric power is applied to carbon electrodes used in the electric furnaces and thermal and mechanical impacts against the electrodes are correspondingly higher; thus, it is required that carbon electrodes be excellent in strength and resistance to spalling.

25 The improvement of carbon electrodes in strength and spalling resistance is one of the most important objectives which manufacturers of carbon electrodes are making efforts to attain; on the other hand, the spalling resistance of the carbon electrodes is essentially conflicting with the strength thereof. It is therefore not easy to obtain carbon electrodes which are satisfactory in both of said properties.

30 In attempts to solve the aforesaid problem, there has heretofore been proposed a process wherein the starting material is specifically limited to needle coke and also a process for impregnating carbon electrodes obtained by baking, with pitch or tar in the molten state

and then re-baking or graphitizing the thus-impregnated carbon electrodes. However, these proposed processes are not expected to improve the resulting carbon electrodes in properties to an extent exceeding a certain limited high extent. For example, when using such carbon electrodes as produced by said known processes, it will be impossible to reduce significantly possible breakage of the electrode bodies, sockets and nipples for the electrode bodies, possible loosening of the screws used in connecting the bodies and nipples possibly caused during the operation of the furnace. In addition, the former one of said known processes is disadvantageous in that such needle coke is usually limited in amount available and the latter one is also disadvantageous in that the rebaking or graphitization after impregnation with pitch in the molten state requires an additional processing stage whereby the total time required to obtain the product is lengthened. In addition, since said proposed processes cannot perfectly eliminate the creation of voids in the carbon electrodes to be obtained nor do they perfectly fill the voids with a suitable material, it would not be expected that they improve the resulting carbon electrodes in properties by more than a predetermined amount.

35 There has recently been reported a process for the production of strengthened carbon electrodes by incorporating carbon fibers in carbonaceous material and baking the resulting mixture to reinforce the carbonaceous material with the carbon fibers. The strengthened carbon electrodes so produced have higher strength than carbon electrodes prepared by filling the voids of carbon electrodes obtained by baking, with pitch or tar and then re-baking the carbon electrodes so filled; since, however, said strengthened carbon electrodes contain the carbon fibers, they have the following disadvantages.

40 (1) The strengthened carbon electrodes are expensive because of the presence of the carbon fibers therein which are obtained by

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heating organic fibers prepared by spinning polyacrylonitrile, rayon or pitch at a relatively low temperature such as 200°—400°C to make them oxygenated or non-fusible (substantially fire-resistant), further heating the thus-oxygenated fibers to about 1400°C to carbonize them and, if desired, still further heating the thus-carbonized fibers to a high temperature up to 3000°C to graphitize them thereby yielding carbon fibers at a very high cost incurred by using the many steps as mentioned above. 5

(2) The carbon fibers are broken into smaller lengths than the original by a violent kneading action under which coke, a binder and the carbon fibers are kneaded together, whereby the carbon fibers so broken do not provide the resulting carbon electrodes with such strength as expected from the original carbon fibers. 10

(3) When baking said kneaded mixture after having been shaped, the carbon fibers will behave themselves in a remarkably different manner than the coke and binder, that is, the former will not change in shape and composition while the latter will decompose and shrink. Therefore, the carbon fibers will not be contained in the baked shaped body in secure adhesion relation with the other ingredients, resulting in remarkably impairing the reinforcing effect of the carbon fibers. As previously mentioned, the carbon electrodes used herein comprise the electrode bodies and nipples for the connection thereof, both of them being made usually of the same carbon. 15

(4) The carbon fibers are unsatisfactory in adhesion to other materials and they may be somewhat improved although to a limited extent, by being surface treated; however, those so surface treated incur an increased production cost. 20

According to the present invention there is provided a process for preparing carbon electrodes, which process comprises the steps of: 25

mixing 100 parts by weight of a carbonaceous material and 0.5—10 parts by weight of non-fusible fibers, 1—25 mm long and 5—15 μ m in diameter, the non-fusible fibers being obtained by oxygenating organic fibers to make at least the surface thereof non-fusible; 30

incorporating the thus-formed mixture with 15—40 parts by weight of a binder; 35

kneading the whole mass; 40

shaping the thus-kneaded mass into green carbon electrodes; and 45

baking the green carbon electrodes at a temperature sufficiently high to carbonize them thereby to prepare the desired carbon electrodes. 50

We have found that if non-fusible, that is oxygenated, organic fibers obtained by heating starting organic fibers to a comparatively low temperature to render them non-fusible or fire-resistant, are kneaded with carbonaceous material such as coke and a binder to form a mixture which was shaped and then baked to obtain a carbon electrode, then the starting organic fibers will not be broken and torn off and their original length is retained even in the step of kneading. The shaped mixture can be baked at 700°—1400°C to form non-graphitized carbon fibers or, if desired further baked at 2400°—3000°C to form graphitized carbon fibers (the term "carbon fibers" being hereinafter intended to mean non-graphitized and graphitized carbon fibers) in the step of baking. The non-fusible organic fibers are relatively similar in composition to the starting or original ones and they evolve therefrom nitrogen, oxygen, hydrogen and the like in the form of low molecular-weight compounds during their decomposition for their carbonization in the baking step. Since the non-fusible organic fibers are carbonized at a carbonizing temperature approximate to that at which the starting carbonaceous material and binder decompose and shrink in the baking step, these three kinds of the materials are closely bonded together into a body after the baking, resulting in the production of a high-strength carbon electrode. 55

Since the tarry material evolved from the non-fusible organic fibers by decomposition thereof is very compatible with that evolved from the binder, co-carbonization yields a baked body or carbon electrode wherein the carbon fibers produced by conversion of the non-fusible fibers in the baking step are securely bonded to the surrounding materials whereby the carbon electrode is further strengthened. 60

The non-fusible organic fibers are produced by a known process which comprises heating organic high molecular-weight fibers obtained by spinning polyacrylonitrile, rayon or pitch, at a comparatively low temperature of not higher than 400°C, e.g. at a temperature of 200—400°C. The use of a temperature higher than 400°C will result in the production of carbon fibers which lack flexibility and tend to be broken in the mixing and kneading steps. 65

Typical of the previously mentioned organic fibers as a starting material for carbon fibers are those of high molecular-weight material having 85% by weight of a polyacrylonitrile which is produced by a usual method and include a homopolymer of acrylonitrile, a copolymer of acrylonitrile with other monomers copolymerizable therewith, and a grafted copolymer of acrylonitrile. 70

The non-fusible polyacrylonitrile fibers may be obtained by heating the starting polyacrylonitrile fibers, without or after immersion thereof in an aqueous solution of an oxide such as potassium permanganate or bichromate for a time sufficient to allow the fibers so change from yellow to brown-yellow in color, to 350°C or lower for at least 10 minutes in an oxidizing atmosphere such as air, a gaseous 75

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5	halogen, oxygen, ozone or gaseous nitric acid. It is desirable that the starting fibers be heated to 200°-300°C for 2-5 hours when they are to be heated in air without the preliminary oxidizing treatment as mentioned above and that the starting fibers be heated to 200°-300°C for 30-60 minutes when they are to be heated in air after the preliminary oxidizing treatment. It is also desirable that when heated in the oxidizing atmosphere, the starting fibers be prevented from shrinking while stretching them under tension so that a stretching ratio (ratio of length after stretch to that (the original) before stretch) of 1 to 1.7 is obtained.	have low intrinsic electrical resistance and low thermal expansion coefficient.	70
10		The optimum amount of the non-fusible organic fibers added varies depending both upon the kind and amount of a carbonaceous material and binder used and upon the properties of the non-fusible fibers. With an increase of the amount of non-fusible fibers added, the resulting carbon electrodes will gradually increase in strength; however, the addition of the fibers in an amount of more than 10% by weight of the carbonaceous material will exhibit no further reinforcing effects and the addition thereof in an amount of 0.5-5% by weight is preferable.	75
15		The particle size of the carbonaceous material such as petroleum coke is preferably in the range of from 12.7 mm to 200 mesh or finer (the mesh standard being the Tyler Sieve Series throughout this specification).	80
20	The previously mentioned pitch fibers may be obtained by melt spinning at 300°-500°C petroleum pitch, coal tar pitch, natural asphalt, a pitch-like material available from certain industrial fields as a py-product, or a specific pitch produced by dry distilling a high molecular-weight material such as polyvinyl chloride or polyacrylonitrile at 300°-400°C to convert the high molecular-weight material	The mixture of the starting materials according to this invention may preferably comprise, by weight, 100 parts of the carbonaceous material such as petroleum coke, 0.5-10 parts of the non-fusible organic fibers such as non-fusible polyacrylonitrile fibers and 15-40 parts of the binder such as coal tar pitch.	85
25	to the specific pitch containing carbon and hydrogen, the specific pitch having a carbon content of 91-95% by weight and an average molecular weight of at least 400.		90
30	The non-fusible pitch fibers may be obtained by oxygenating the starting pitch fibers at 300°C or lower for at least 10 minutes in an oxidizing atmosphere of air, oxygen, ozone or nitrogen oxides. It is desirable that when the starting pitch fibers are to be oxygenated in air, they should be treated with ozone at 40°-70°C for 2-5 hours before being heated in air at 200°-260°C for 5-20 hours.	As previously mentioned, according to this invention, the non-fusible organic fibers are not broken with the original size being retained in the step of kneading with carbonaceous material and a binder to form a mixture, and the non-fusible organic fibers and the carbonaceous material in particulate form may be blended together usually for 60 minutes or less. If the blending is effected not by a low speed mixer, such as Warner, Ribbon or Drum type one, having rotation vanes capable of rotating at a peripheral velocity of as low as 20-60 metres/minute, but by a high speed mixer, such as Henschel mixer manufactured by Henschel Company in West Germany, having rotation vanes capable of rotating at a peripheral velocity of as high as 5-50 metres/second, then the mixing will be completed to a satisfactory extent in 5-15 minutes. The packing density varies depending on the kind and particle size of the starting materials and it is difficult to uniformly disperse and mix the rough particles and fine particles of the carbonaceous material with the non-fusible fibers by the use of such conventional low speed mixer. Because of this, the resulting carbon electrodes will tend to have cavities and fine cracks therein thereby making it difficult to obtain compact and homogeneous carbon electrodes. To solve this problem, it is desirable to employ such a high speed mixer as mentioned above. It has been confirmed that the use of the high speed mixer having agitation vanes rotatable at a peripheral velocity of 5-50 metres/second enables the rough and fine particles and the fibers to be mixed together in a fluid-	95
35	The previously mentioned rayon fibers may preferably be regenerated cellulose fibers such as viscose or acetate rayon fibers wherein the rayon has a polymerization degree of at least 450.		100
40	The non-fusible rayon fibers may be obtained by heating the starting rayon fibers to 200°-400°C for 10-20 hours in a non-oxidizing atmosphere of nitrogen, argon or helium or by heating the same to 200°-400°C for 1-10 hours in an atmosphere containing 1-20% by volume of hydrogen chloride together with nitrogen, argon or helium.		105
45	Among the non-fusible fibers which may be used in the process of the invention, those of polyacrylonitrile origin are particularly preferred in view of their excellent strength and flexibility. They have a tensile strength of 10-20 Kg/mm ² which is 2-10 times higher than those of non-polyacrylonitrile origin and tend less to be broken or torn off particularly in the mixing and kneading steps. In addition, as compared with the non-fusible non-polyacrylonitrile fibers, the non-fusible polyacrylonitrile fibers are in satisfactory graphitized form and are advantageous in that the graphitized carbon electrodes containing them		110
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ized state thereby obtaining a thoroughly mixed and homogeneous mixture. The use of a peripheral velocity of lower than 5 metres/second will not exhibit a satisfactory dispersion of the starting materials, while the use of a peripheral velocity of higher than 50 metres/second will undesirably pulverize the rough particles and the fibers.

The mixed particles obtained by mixing and dispersing the particulate carbonaceous material and the non-fusible fibers, for example by the use of the high speed mixer, are incorporated with the binder to form a mixture which is kneaded, shaped and baked for carbonization or graphitization. The amount of the binder so incorporated may be decreased by 3-10% by weight of the whole mass. Thus in the heating step, the volatile ingredients are evolved in a small amount, and the densification of the resulting product and the improvement in the internal structure of the product may be easily attained. The graphite articles obtained by the process of this invention using the high speed mixing and kneading are 10-20% higher in bulk specific gravity, 15-30% lower in intrinsic resistance and 10-50% higher in bending strength, than the conventional graphite articles. High speed mixers which may preferably be used for high speed mixing, include Henschel mixers and super precision high speed mixers; it is desirable to select the shape of the agitating vanes of the mixers for further better mixing. The mixture so formed is shaped and baked to carbonize the non-fusible fibers simultaneously with the carbonization of the carbonaceous material and binder. In the baking step, the non-fusible organic fibers decompose at a temperature approximately that at which the carbonaceous material and binder decompose while the tarry compounds evolved from these three kinds of the materials are soluble in each other and carbonized, whereby the carbon fibers produced by the carbonization of the non-fusible fibers are securely bonded to the surrounding material in the shaped body, thus yielding high-strength carbon electrodes with the use of less heat and labour.

This invention will be better understood by the following non-limitative Examples where-

in all parts are by weight unless otherwise specified.

Examples 1-2, and Comparative example 1. 55

Non-fusible organic fibers, 5 mm long and 10 μ m in diameter, prepared by heating organic fibers consisting of a bundle of 6000 1.5-denier monofilaments of a resin containing 93 wt % polyacrylonitrile (the monofilament having a dry strength of 5 grams/denier and an elongation of 16%), in air at 200°-300°C for 3 hours while stretching the organic fibers at a stretching ratio of 1.2 to form non-fusible fibers in a bundle which were then cut into the pieces, 5 mm long and 10 μ m in diameter. Each of 0.5 parts and 1 part of the non-fusible fibers so prepared was blended with 35 parts of pulverized and sieved petroleum coke having a 14-48 mesh size, 60 parts of the same pulverized coke having not greater than a 200 mesh size and 5 parts of pulverized artificial graphite. The whole mass was mixed on a Henschel mixer having agitating vanes rotating at a peripheral speed of 40 m/sec. for 10 minutes and transferred to a Warner type kneader where it was incorporated with 26 parts of medium pitch (softening point, 87°C) and then kneaded at 150°C for one hour. The mixtures so kneaded were shaped respectively into rods, 10" in diameter and 1500 mm long, by the use of an extrusion press. The rods so obtained were placed in a baking furnace where they were baked at a temperature which was gradually raised from ambient temperature to 700°C in 220 hours, and they were then transferred into an electric graphitizing furnace where they were heated up to about 3000°C for graphitization. The thus-graphitized rods were cooled and cut into cubes, 20 mm \times 20 mm \times 100 mm, as test pieces. The test pieces were tested for their properties which are shown in Table 1. 60

For comparison, graphitized rods were prepared in the same manner as in Examples 1 and 2 except that the non-fusible organic fibers were not used and they were then cut into cubes having the same size as those in Examples 1 and 2. The comparative test pieces were subjected to the same test as above with the result being also shown in Table 1. 65

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TABLE 1

	Example 1	Example 2	Comparative example 1
Amount of non-fusible fibers added	0.5	1	0
Bulk specific gravity	1.73	1.73	1.72
Bending strength (Kg/cm ²)	250	270	220
Specific resistance ($\times 10^{-5}$ Ω.cm)	67	63	74

From Table 1 it is apparent that the addition of the non-fusible fibers resulted in the production of carbon electrodes having improved properties.

Example 3, and Comparative examples 2-3.
 One part of non-fusible organic fibers, 10 mm long and 10 μ m in diameter, prepared in the same manner as in Example 1 or 2, and one part of carbon fibers, 10 mm long and 5 μ m in diameter, prepared by heating the same non-fusible organic fibers as above to 1300°C in a nitrogen gas atmosphere for 10 seconds and then cutting the resulting carbon fibers, were each added to a blend of 35 parts of pulverized and sieved petroleum coke having a 14-48 mesh size, 60 parts of the same coke having a size of 200 mesh or finer and 5 parts of pulverized artificial graphite, to form a mixture which was mixed for 20 minutes on a Henschel mixer having agitating vanes rotating at a peripheral velocity of 40 m/sec., incorporated with 26 parts of medium pitch (softening point, 87°C) and kneaded at 150°C for one hour. The kneaded mixtures so obtained were subsequently treated in the same manner as in Example 1 to prepare test pieces of the same size. The test pieces so prepared were tested for their properties in the same manner as previously mentioned and the results are shown in Table 2.

In Comparative example 2 the procedure of Example 3 was followed except that carbon

fibers were substituted for the non-fusible organic fibers and in Comparative example 3 the procedure of Example 3 was repeated except that none of the carbon fibers and non-fusible organic fibers were used, to obtain in each Comparative example a graphite product from which were made test graphite pieces having the same size as those in Example 3. The test pieces so made were tested for their properties with the results being indicated in Table 2.

From Table 2 it is appreciated that the addition of the non-fusible fibers resulted in the production of carbon electrodes having improved properties.

Comparative example 4.
 The starting materials having the same composition as those used in Example 3 were mixed and kneaded together for 50 minutes in a low speed mixer the agitating vanes of which rotated at a peripheral velocity of 1 m/sec., with the result that a satisfactory mixing and kneading was not obtained. Therefore, the subsequent operations were not carried out.

The carbon products of this invention may be used not only as carbon electrodes but also as nipples without any troubles even though they are those which were prepared without being subjected to additional treatments such as pitch impregnation and re-baking or graphitization after baking unlike the conventional carbon products.

TABLE 2

	Example 3	Comparative example 2	Comparative example 3
Kind of fibers added	Non-fusible fibers	Carbon fibers	None
Amount of fibers added	1	1	0
Bulk specific gravity of test pieces	1.71	1.71	1.72
Bending strength of test pieces (Kg/cm ²)	260	240	215
Specific resistance of test pieces ($\times 10^{-5} \Omega \cdot \text{cm}$)	64	75	73

Examples 4—5, and
Comparison example 5.

5 Non-fusible fibers (tensile strength, 19 Kg/mm²) consisting of 18000 polyacrylonitrile filaments having a 10 μm diameter, prepared in the same manner as in Example 1, were passed through a bath containing medium pitch softening point (87°C) in molten state and passed between two rollers to remove the excess pitch carried on the fibers thereby obtaining a bundle of the fibers coated with the pitch in an amount of about 300% of the original fibers. The thus coated fibers were cooled and cut into smaller fibers of about 10 mm in length.

10 15 One part of and 3 parts (in each case, the amount of fibers only, except for the amount of pitch coated) of the smaller fibers so obtained were each added to a mixture containing 50 parts of petroleum coke having a

25 particle size of 0.50—0.15 mm and 50 parts of the same coke the 60% of which is not larger than 0.074 mm in particle size to form a mixture which was then blended. The two mixtures so blended were each incorporated with 32 parts of medium pitch (softening point 87°C). The whole mass was kneaded for 50 minutes and then extrusion molded into shaped bodies having a size of 10" diameter \times 1500 mm which were baked, impregnated with pitch and rebaked (the impregnation and rebaking being effected twice, respectively) at 2700°C or higher thereby obtained graphitized carbon electrodes. The properties of the products are shown in Table 3.

30 35 For comparison, the procedure of Example 4 was followed except that the non-fusible fibers are not added to obtain comparative graphitized carbon electrodes. The properties of the products are shown in Table 3.

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TABLE 3

	Example 4	Example 5	Comparative example 5
Amount of fibers added (%)	1	3	0
Bulk specific gravity	1.73	1.71	1.75
Specific resistance ($\times 10^{-5} \Omega \cdot \text{cm}$)	41	42	42
Bending strength (Kg/cm ²)	270	290	250
Modulus of elasticity (Kg/mm ²)	1470	1480	1450
Thermal expansion coefficient ($10^{-6}/^\circ\text{C}$)	1.00	0.90	1.10

Notes to Table 4:

Furnace capacity 50 tonnes, Transformer capacity 22 megavolt amperes,

Secondary current 40 kiloamperes, Load applied 20 megawatts.

The results of practical tests on the nipples are as follows:

Percentage of nipples broken: 1 in Example 4

0 in Example 5

5 in Comparative example 5

Example 6 and
Comparative example 6.

5 One part (the amount of fibers only, except for the pitch coated) of the same smaller non-fusible fibers, 10 mm long, as those of Example 4 was added to a blend of 50 parts of petroleum coke having a particle size of 1—0.50 mm and 50 parts of the same coke wherein 60% of the coke has a particle size of not larger than 0.074 mm to form a mixture which was mixed, incorporated with 28

parts of medium pitch (softening point 87°C) and kneaded for 50 minutes. The whole mass so kneaded was extrusion molded into shaped bodies having a size of 20" diameter×6' which were baked at 800°C and then further baked at 2700°C or higher thereby to obtain graphitized carbon electrodes. The properties of the products are indicated in Table 4.

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For comparison, the procedure of Example 6 was followed except that the non-fusible fibers are not added. The properties of the products are shown in Table 4.

TABLE 4

	Example 6	Comparative example 6
Amount of fibers added (%)	1	0
Bulk specific gravity	1.58	1.60
Specific resistance ($\times 10^{-5} \Omega \cdot \text{cm}$)	75	80
Bending strength (Kg/cm ²)	110	90
Modulus of elasticity (Kg/mm ²)	610	590
Thermal expansion coefficient ($\times 10^{-6}/^\circ\text{C}$)	1.2	1.3

The results of practical tests are as follows:

Percentage of electrode pole sockets broken: zero in Example 6

3 in Comparative example 6

WHAT WE CLAIM IS:—

1. A process for preparing carbon electrodes, which process comprises the steps of:

30 mixing 100 parts by weight of a carbonaceous material and 0.5—10 parts by weight of non-fusible fibers, 1—25 mm long and 5—15 μm in diameter, the non-fusible fibers being obtained by oxygenating organic fibers

to make at least the surface thereof non-fusible;

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incorporating the thus-formed mixture with

15—40 parts by weight of a binder;

kneading the whole mass;

shaping the thus-kneaded mass into green carbon electrodes; and

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baking the green carbon electrodes at a temperature sufficiently high to carbonise them

thereby to prepare the desired carbon electrodes.

2. A process according to claim 1, wherein the oxygenating is effected at a temperature of 200—400°C.

5 3. A process according to claim 1 or 2, wherein the baking is effected at a temperature of 700—1400°C.

10 4. A process according to claim 1 or 2, wherein the baking is effected at a temperature sufficiently high to graphitise the green carbon electrodes.

15 5. A process according to claim 4, wherein the baking is effected at a temperature of 2400—3000°C.

6. A process according to any preceding claim, wherein the carbonaceous material and the non-fusible fibers are mixed for 5 to 15 minutes in a high speed mixer having vanes capable of rotating at a peripheral velocity of 5 to 50 metres/second.

20 7. A process according to any preceding claim, wherein the carbonaceous material comprises coke.

8. A process according to any preceding claim, wherein the organic fibers are comprised of polyacrylonitrile.

25 9. A process according to any one of claims 1 to 7, wherein the organic fibers are comprised of pitch.

10. A process according to any one of claims 1 to 7, wherein the organic fibers are comprised of rayon.

30 11. A process according to any preceding claim, wherein 0.5—5 parts by weight of non-fusible fibers are mixed with the carbonaceous material.

12. A process according to any preceding claim, wherein the binder is comprised of pitch.

35 13. A process according to any one of claims 1 to 11, wherein the binder is comprised of tar.

14. A process according to any one of claims 1 to 11, wherein the binder is comprised of a synthetic resin.

40 15. A process according to any preceding claim, wherein the non-fusible fibers being in an amount of 1 to 3 parts by weight and are provided with a coating of pitch before being cut to length, the amount of the coated fibers being determined in the absence of the coating.

16. A process according to claim 15, wherein the proportion of binder is from 28 to 32 parts by weight.

17. A process for preparing carbon electrodes according to claim 1 and substantially as hereinbefore described.

18. A process for preparing carbon electrodes according to claim 1 and substantially as described in any one of the foregoing Examples.

19. Carbon electrodes made by a process according to any preceding claim.

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